
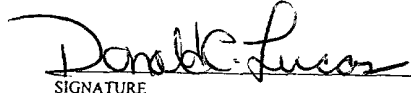


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|---|--|---|--|---|--|
| FORM PTO-1390 (REV 11-98) | | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE | | ATTORNEY'S DOCKET NUMBER | |
| TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 | | | | E-1024 | |
| | | | | U.S. APPLICATION NO. (If known, see 37 CFR 1.5) | |
| INTERNATIONAL APPLICATION NO. PCT/NO99/00336 | | INTERNATIONAL FILING DATE 8 November 1999 | | PRIORITY DATE CLAIMED 9 November 1998 | |
| TITLE OF INVENTION RESIN COMPOSITIONS, METHOD OF PRODUCING RESIN COMPOSITIONS AND FILLER BLENDS FOR USE IN RESIN COMPOSITIONS | | | | | |
| APPLICANT(S) FOR DO/EO/US DANIELSSEN, Tore; LINNEBO, Anne; and SANDELIN, Bjorn | | | | | |
| Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information. | | | | | |
| <p>1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371</p> <p>2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1)</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p style="margin-left: 20px;">a. <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau)</p> <p style="margin-left: 20px;">b. <input type="checkbox"/> has been transmitted by the International Bureau.</p> <p style="margin-left: 20px;">c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</p> <p>6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2))</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p style="margin-left: 20px;">a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</p> <p style="margin-left: 20px;">b. <input type="checkbox"/> have been transmitted by the International Bureau.</p> <p style="margin-left: 20px;">c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</p> <p style="margin-left: 20px;">d. <input type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</p> <p>Items 11. to 16. below concern document(s) or information included:</p> <p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</p> <p>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</p> <p>13. <input type="checkbox"/> A FIRST preliminary amendment</p> <p style="margin-left: 20px;"><input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter.</p> <p>16. <input checked="" type="checkbox"/> Other items or information: Form PTO-2038</p> | | | | | |

| U.S. APPLICATION NO. (If known, see 37 CFR 1.53) | | INTERNATIONAL APPLICATION NO. | | ATTORNEY'S DOCKET NUMBER | |
|---|--------------|-------------------------------|------------|--|----------|
| 097830860 | | PCT/NO99/00336 | | E-1024 | |
| 17. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) : Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$1,000 and International Search Report not prepared by the EPO or JPO \$840.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$840.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$760.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$670.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) \$96.00 ENTER APPROPRIATE BASIC FEE AMOUNT = | | | | \$ 1,000 | |
| Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)). | | | | \$ | |
| CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | | |
| Total claims | - 20 = | | X \$18.00 | \$ | |
| Independent claims | - 3 = | | X \$78.00 | \$ | |
| MULTIPLE DEPENDENT CLAIM(S) (if applicable) | | | + \$260.00 | \$ | |
| TOTAL OF ABOVE CALCULATIONS = | | | | \$ 1,000 | |
| Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) | | | | \$ | |
| SUBTOTAL = | | | | \$ | |
| Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)). | | | | \$ | |
| TOTAL NATIONAL FEE = | | | | \$ 1,000 | |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property | | | | \$ | |
| TOTAL FEES ENCLOSED = | | | | \$ 1,000 | |
| Form PTO-2038 | | | | Amount to be: refunded | \$ |
| | | | | charged | \$ 1,000 |
| a. <input checked="" type="checkbox"/> A check in the amount of \$ <u>1,000</u> to cover the above fees is enclosed. XXXXXX Form PTO-2038 | | | | | |
| b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. | | | | | |
| c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>02-2275</u> . A duplicate copy of this sheet is enclosed. | | | | | |
| NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status. | | | | | |
| SEND ALL CORRESPONDENCE TO: Donald C. Lucas BIERMAN, MUSERLIAN AND LUCAS 600 Third Avenue New York, NY 10016 | | | | | |
|  20311 <small>PATENT TRADEMARK OFFICE</small> | | | |  <small>SIGNATURE</small> | |
| | | | | <u>Donald C. Lucas</u> <small>NAME</small> | |
| | | | | <u>31,275</u> <small>REGISTRATION NUMBER</small> | |

09/830860

JC18 Rec'd PCT/PTO 3 0 APR 2001

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Date of Deposit: April 30, 2001

I hereby certify that this correspondence is being deposited
with the United States Postal Service "EXPRESS MAIL POST OFFICE TO
ADDRESSEE" Service under 37 CFR 1.10 on the date indicated above
and is addressed to Asst. Commissioner for Patents, Washington,
D.C. 20231.

D. Nakonezny

Title of Invention

Resin compositions, method of producing resin compositions and filler blends for use in resin compositions.

5 Technical Field

The present invention relates to new and improved resin compositions and more particularly to thermoplastic resin compositions such as polyolefines, polyvinylchloride and polyamide, and to a method for the production of resin compositions. The invention further relates to a filler blend for use in the
10 production of resin compositions.

Background Art

It is well known to produce polyolefines such as polypropylene compound containing functional fillers such as fine particulate talc to increase the
15 stiffness of the final polypropylene product.

Talc is hydrated magnesium silicate with the theoretical formula $3\text{MgO} \cdot 4\text{SiO}_4 \cdot \text{H}_2\text{O}$ and consists of magnesiumhydroxide sandwiched between two sheets of silica.
20

When adding other fillers in addition to talc in order to improve other properties, such as for example impact strength, it has, however, been found that the stiffness obtained by using talc alone as a filler is substantially reduced when adding a second filler for increasing the impact strength. It has
25 therefore not been possible to produce polypropylene products with both a high stiffness and a high impact strength. High stiffness and high impact strength is particularly important in some polypropylene products such as for example car bumpers. The same is true for other thermoplastic resin products.

30

The term thermoplastic resin used in the specification and claims includes not only thermoplastic resins per se, but also mixtures thereof, as well as a blend

of thermoplastic resins with other materials such as an elastomer like nitrile rubber. The so-called thermoplastic rubbers, thermoplastic elastomers are also included in the definition of thermoplastic resin. Thermoplastic resins per se includes polyolefines, polystyrene, polyesters, ABS copolymers, polyvinyl chloride (PVC), unplasticized polyvinyl chloride (UPVC), polyamide, acrylic polymers, polycarbonate polymers, polysulfone polymers and others.

It is known from US patent No. 4,722,952 that the addition of microsilica to polyvinylchloride, improves the impact strength of polyvinylchloride used for the production of electrical conduits. For such products the stiffness is of no importance. On the contrary, high stiffness is not desired for electrical conduits.

The term microsilica used in the specification and claims is particulate amorphous SiO_2 obtained from a process in which silica is reduced and the reduction product is oxidized in vapor phase to form amorphous silica. Microsilica may contain at least 70 % by weight silica (SiO_2) and have a specific density of 2,1 - 2,3 g/cm^3 and a surface area at 15 - 30 m^2/g . The primary particles are substantially spherical. The primary particles have an average size of about 0,15 μm . Microsilica is preferably obtained as a co-product in the production of silicon or silicon alloys in electric reduction furnaces. In these processes large quantities of silica are formed as SiO_2 . The SiO_2 is recovered in conventional manner using filter or other collection apparatus. For the purpose of the present invention the term microsilica also shall be understood to include fly-ash, and more particularly fly-ash particles of substantial spherical shape having a particle size below 10 microns.

Disclosure of Invention

It is an object of the present invention to provide thermoplastic resins having both high stiffness and high impact strength.

According to a first aspect, the present invention thus relates to thermoplastic resin compositions, particularly polyolefines, polyvinylchloride and polyamide, characterized in that the thermoplastic resin compositions contains between 3 and 400 % by weight of filler based on the weight of the resin, said filler comprising talc and microsilica where the weight ratio between talc and microsilica is between 15:1 and 1:15.

According to a preferred embodiment the weight ratio of talc and microsilica is between 6:1 and 1:5.

According to a second aspect the present invention relates to a method for the production of thermoplastic resin composition, particularly polyolefines, polyvinylchloride and polyamide, which method being characterized in that talc and microsilica is added to thermoplastic resin in a total amount between 3 and 400 % by weight based on the weight of thermoplastic resin and where the weight ratio between talc and microsilica is kept between 15:1 and 1:15, whereafter the mixture is formed into a thermoplastic resin product or compound.

According to a preferred embodiment of the method of the present invention talc and microsilica are added to the thermoplastic resin as a mixture of talc and microsilica.

The compounding of the thermoplastic resin can be done using conventional processes like extrusion, calendering, injection moulding and others.

According to a third aspect, the present invention relates to a filler blend for use in thermoplastic resins, particularly polyolefines, polyvinylchloride and polyamide, wherein the filler blend contains talc and microsilica in a weight ratio between 15:1 and 1:15, and particularly between 6:1 and 1:5.

It has surprisingly been found that the combined use of talc and microsilica as fillers in thermoplastic resins, particularly in polyolefines, polyvinylchloride and polyamide, give final products having both high stiffness and high impact strength.

5

EXAMPLE 1

A non-filled polypropylene copolymer "BA 202E" supplied by Borealis was extruded in a compounding extruder with addition of a filler blend consisting of talc supplied by Mondo Minerals OY and microsilica supplied by Elkem ASA. The weight ratio between talc and microsilica in the filler blend was 2:1 and tests were run with addition of 5, 10 and 19 % by weight of the filler blend based on the weight of the polypropylene copolymer. The stiffness of the extruded polypropylene was measured as tensile modulus according to ISO 527 and the impact strength of the extruded polypropylene was measured as notched charpy impact strength according to ISO 179/1A.

For comparison purposes the polypropylene copolymer was extruded in the compounding extruder with no addition of filler and with the addition 5, 10 and 18 % by weight of talc and with 5 and 10 % by weight of microsilica. Also for these comparative tests the stiffness and the impact strength were measured as stated above. The resulting stiffness and impact strength are shown in figure 1 and figure 2 respectively.

As can be seen from figure 1 and 2, the impact strength of the polypropylene containing both talc and microsilica is much higher than for the polypropylene containing only talc and only slightly lower than for the polypropylene containing only microsilica as a filler. The stiffness of the polypropylene containing both talc and microsilica is much higher than for polypropylene containing only microsilica as a filler and only slightly lower than for polypropylene containing only talc as a filler. The use of a blend of talc and

microsilica thus surprisingly gives a polypropylene having both a high stiffness and a high impact strength.

EXAMPLE 2

A non-filled high density polyethylene (HDPE) copolymer "HDPE HE 2467-BL" supplied by Borealis was extruded in a compounding extruder with addition of a filler blend consisting of talc supplied by Mondo Minerals OY and microsilica supplied by Elkem ASA. The weight ratio between talc and microsilica in the filler blend was 2:1 and the test was run with addition of 10 % by weight of the filler blend based on the weight of the HDPE copolymer. The stiffness of the extruded HDPE was measured as tensile modulus according to ISO 527 and the impact strength of the extruded HDPE was measured as notched charpy impact strength according to ISO 179/1A.

For comparison purposes the HDPE copolymer was extruded in the compounding extruder with no addition of filler, with the addition 10 % by weight of talc and with addition of 10 % by weight of microsilica. Also for these comparative tests the stiffness and the impact strength were measured as stated above. The resulting stiffness and impact strength are shown in table 1.

Table 1

| Material | Tensile Modulus (MPa) | Impact Strength (kJ/m ²) |
|--------------------------|-----------------------|--------------------------------------|
| HDPE nonfilled | 850 | 13.6 |
| HDPE + 10 % talc | 1160 | 18.0 |
| HDPE + 10 % microsilica | 880 | 27.6 |
| HDPE + 10 % filler blend | 1070 | 22.3 |

As can be seen from table 1, the impact strength of the HDPE containing both talc and microsilica is higher than for the HDPE containing only talc, but lower than for the HDPE containing only microsilica as a filler. The stiffness of the HDPE containing both talc and microsilica is much higher than for HDPE containing only microsilica as a filler and only slightly lower than for HDPE containing only talc as a filler. The use of a blend of talc and microsilica thus surprisingly resulting in a HDPE having both a high stiffness and a high impact strength.

10 EXAMPLE 3

A non-filled polyvinylchloride (PVC) polymer was calendered with additon of a filler blend consisting of talc supplied by Mondo Minerals OY and microsilica supplied by Elkem ASA. The weight ratio between talc and microsilica in the filler blend was 2:1 in one run and 1:2 in another run, and the tests were run with addition of 5 % by weight of the filler blend based on the weight of PVC polymer. The stiffness of the calendered PVC was measured as tensile modulus according to ISO 527 and the impact strength of the calendered PVC was measured as notched charpy impact strength according to ISO 179/1A.

For comparison purposes the PVC polymer was calendered with no addition of filler, with addition of 5 % by weight of talc and with addition of 5 % by weight of microsilica. Also for these comparative tests the stiffness and the impact strength were measured as stated above. The resulting stiffness and impact strength are shown in table 2.

Table 2

| Material | Tensile Modulus (MPa) | Impact Strength (kJ/m ²) |
|---|-----------------------|--------------------------------------|
| PVC nonfilled | 2916 | 6.5 |
| PVC + 5 % talc | 3484 | 5.4 |
| PVC + 5 % microsilica | 3010 | 8.5 |
| PVC + 5 % filler blend talc/microsilica 2:1 | 3360 | 5.1 |
| PVC + 5 % filler blend talc/microsilica 1:2 | 3167 | 7.9 |

As can be seen from table 2, the impact strength of PVC containing talc and microsilica in a ratio of 2:1 is about the same as for the PVC containing only talc, but lower than for PVC containing only microsilica as a filler. For PVC containing talc and microsilica in a ratio of 1:2 it can be seen that the impact strength is higher than for PVC containing talc and microsilica in a ratio of 2:1 and almost as high as for PVC containing only microsilica. The stiffness of the PVC containing talc and microsilica in a ratio of 2:1 is much higher than for PVC containing only microsilica as a filler and only slightly lower than for PVC containing only talc as a filler. For PVC containing talc and microsilica in a ratio of 1:2 it will be seen that the tensile modulus is still higher than for PVC containing only microsilica. The use of a blend of talc and microsilica thus surprisingly gives a PVC having both a high stiffness and a high impact strength.

EXAMPLE 4

A non filled polyamide (PA) polymer, "PA6 Ultramid B35" delivered by BASF was extruded in a compounding extruder with addition of a filler blend consisting of talc supplied by Mondo Minerals OY and microsilica supplied by

Elkem ASA. The addition of filler blend was 10 % by weight of polymer. The weight ratio between talc and microsilica in the filler blend in a first test was 1:1 and 1:2 in a the second test. The stiffness of the extruded PA was measured as tensile modulus according to ISO 527 and the impact strength of the extruded PA was measured as notched charpy impact strength according to ISA 179/1A.

For comparison purposes the PA copolymer was extruded in the compounding extruder with no addition of filler, with the addition 10 % by weight of talc and with addition of 10 % by weight of microsilica. Also for these comparative tests the stiffness and the impact strength were measured as stated above. The resulting stiffness and impact strength are shown in table 3.

Table 3

| Material | Tensile Modulus (MPa) | Impact Strength (kJ/m ²) |
|---|-----------------------|--------------------------------------|
| PA nonfilled | 700 | Non-break |
| PA + 10 % talc | 1430 | 10.6 |
| PA + 10 % microsilica | 890 | 33.2 |
| PA + 10 % filler blend talc/microsilica 1:1 | 1210 | 16.3 |
| PA + 10 % filler blend talc/microsilica 1:2 | 1120 | 19.7 |

As can be seen from table 3, the impact strength of the PA containing both talc and microsilica is much higher than for the PA containing only talc, but lower than for the PA containing only microsilica as a filler. It can also be seen that the impact strength increases with increasing amount of microsilica in the filler blend. The stiffness of the PA containing both talc and microsilica is

much higher than for PA containing only microsilica, but the stiffness is slightly reduced when the microsilica content in the filler blend is increased.

CLAIMS

- 5 1. Thermoplastic resins compositions, particularly polyolefines, polyvinylchloride and polyamide, characterized in that the thermoplastic resin compositions contain between 3 and 400 % by weight of filler based on the weight of the resin, said filler comprising talc and microsilica where the weight ratio between talc and microsilica is between
10 15:1 and 1:15.
2. Thermoplastic resins according to claim 1, characterized in that the weight ratio of talc and microsilica is between 6:1 and 1:5.
- 15 3. A method for the production of thermoplastic resin composition, particularly polyolefines, polyvinylchloride and polyamide, characterized in that talc and microsilica is added to the thermoplastic resin in a total amount between 3 and 400 % by weight based on the weight of thermoplastic resin and where the weight ratio between talc and microsilica is
20 kept between 15:1 and 1:15, whereafter the mixture is formed to a thermoplastic resin product or compound.
4. A method according to claim 3, characterized in that talc and microsilica are added to the thermoplastic resin as a mixture of talc and
25 microsilica.
5. A method according to claim 2, characterized in that talc and microsilica are added separately to the thermoplastic resin.
- 30 6. A filler blend for use in thermoplastic resin compositions, particularly polyolefines, polyvinylchloride and polyamide, characterized in

that the filler blend contains talc and microsilica in a weight ratio between 15:1 and 1:15.

7. A filler blend according to claim 6, characterized in that the
5 filler blend contains talc and microsilica in a weight ratio between 6:1 and 1:5.

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Stiffness of talc/EMS in PP copolymer

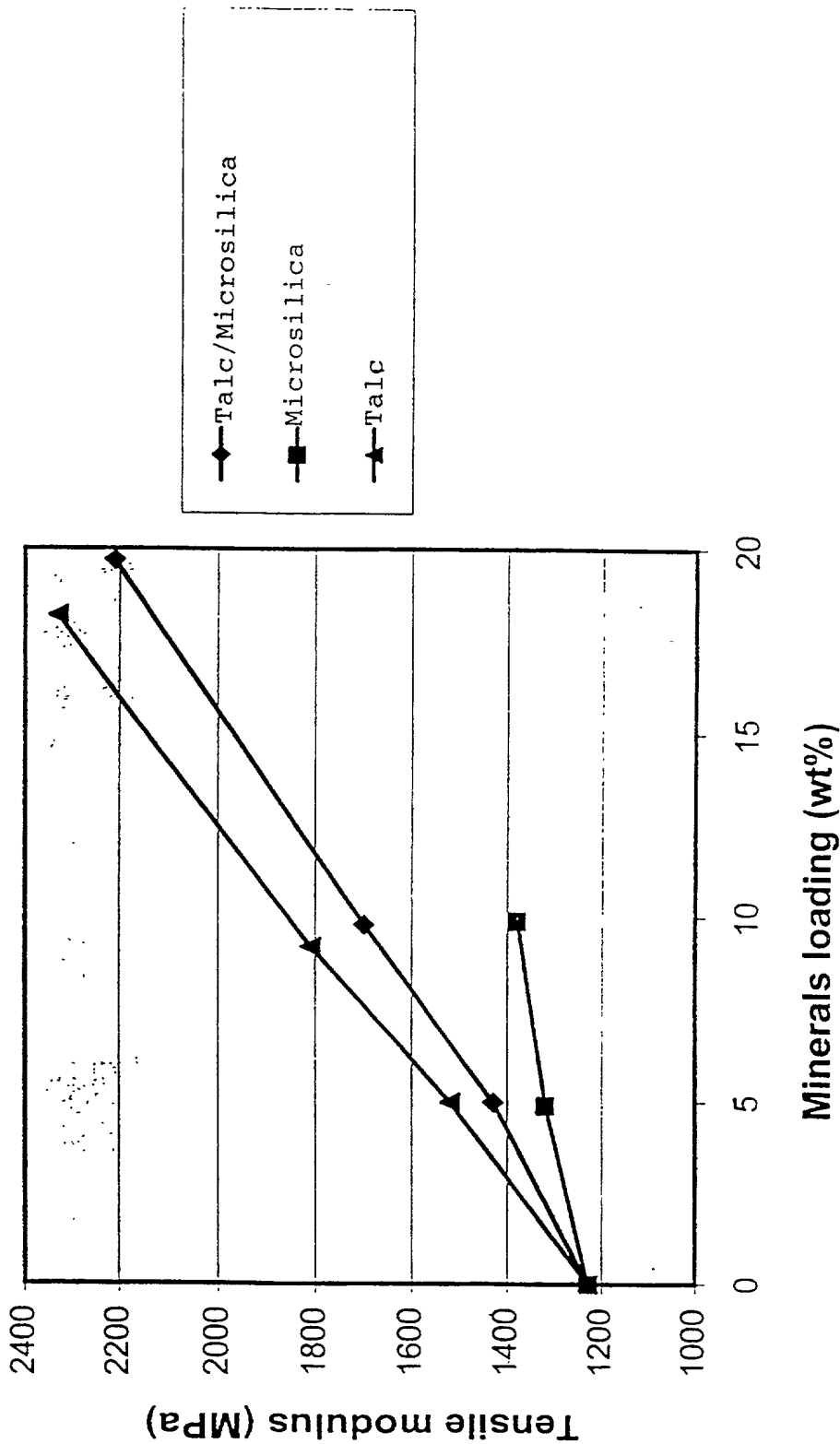


FIGURE 1

Impact strength of talc/EMS in PP copolymer

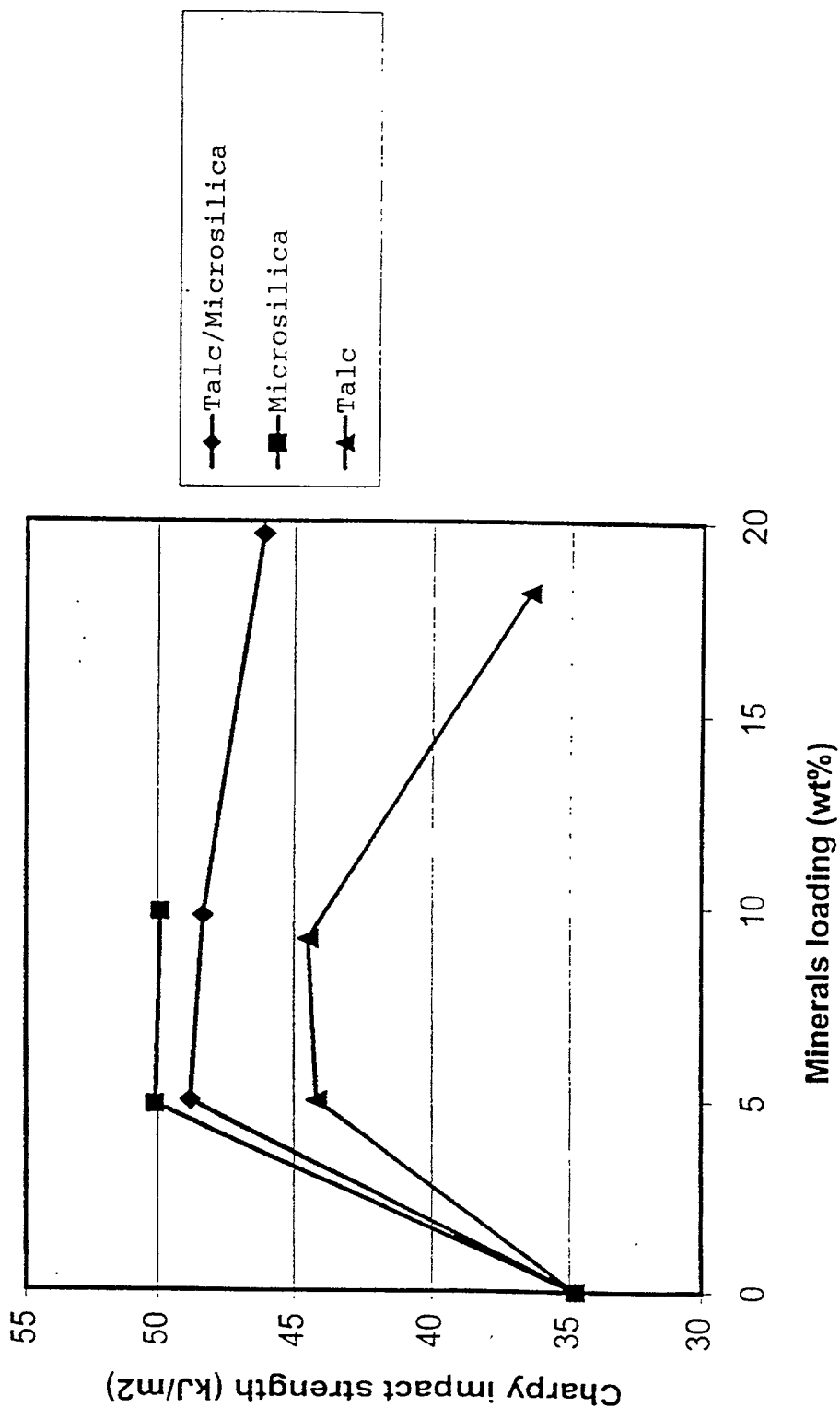


FIGURE 2

DECLARATION AND POWER OF ATTORNEY

E-1024

US

#4

Each below-named inventor hereby declares and says that:

My residence, post office address and citizenship are as stated below beneath my name; I verily believe I am the original, first and sole inventor (if only one name is listed below) or a joint inventor (if plural inventors are named below) of the invention titled: **RESIN COMPOSITIONS, METHOD OF PRODUCING RESIN COMPOSITIONS AND FILLER BLENDS FOR USE IN RESIN COMPOSITIONS**

which is described and claimed in the attached application, or Serial No. _____ filed _____ as amended to date; I have reviewed and understand the contents of the specification including the claims with all the above-mentioned amendments thereto, if any. I acknowledge my duty to disclose information of which I am aware which is material to the patentability of this application in accordance with 37 CFR 1.56; and, if the benefit of 35 U.S.C. 120 is claimed below, as to subject matter of the claims not disclosed in any prior U.S. application in accordance with 35 U.S.C. 112, I acknowledge my duty to disclose material information which became known to me between the filing date of said prior U.S. application and this application and is material to the patentability of this application as defined in 37 CFR 1.56; the benefit of 35 U.S.C. 119(e), or 120 is claimed for

SERIAL NO

FILED

STATUS

I claim the foreign priority benefits under 35 U.S.C. 119 of foreign application(s) for patent or inventor's certificate(s) filed less than 12 months prior to the filing of the application, or less than 12 months before the application(s) for which the above benefit of 35 U.S.C. 120 is claimed as follows:

COUNTRY

SERIAL NO.

FILING DATE

PCT

PCT/NO99/00336

November 8, 1999

Norway

19985211

November 9, 1998

and I have identified any foreign application(s) for patent or inventor's certificate(s) having a filing date before the earliest of the application(s) for which priority is claimed, or the present application, as follows:

COUNTRY

SERIAL NO.

FILING DATE

BIERMAN, MUSERLIAN and LUCAS LLP, Customer No. 20311, Reg. No. 18,818; JORDAN B. BIERMAN, Reg. No. 18,629; CHARLES A. MUSERLIAN, Reg. No. 19,683; and DONALD C. LUCAS, Reg. No. 31,275; all of 600 Third Avenue, New York, New York 10016, Telephone (212) 661-8000, are hereby

It is declared by undersigned that all statements made herein of undersigned's own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S. Code 1001, and that such willful false statements may jeopardize the validity of this application or any other patent issuing thereon.

RESIDENCE AND POST OFFICE ADDRESS

[illegible]